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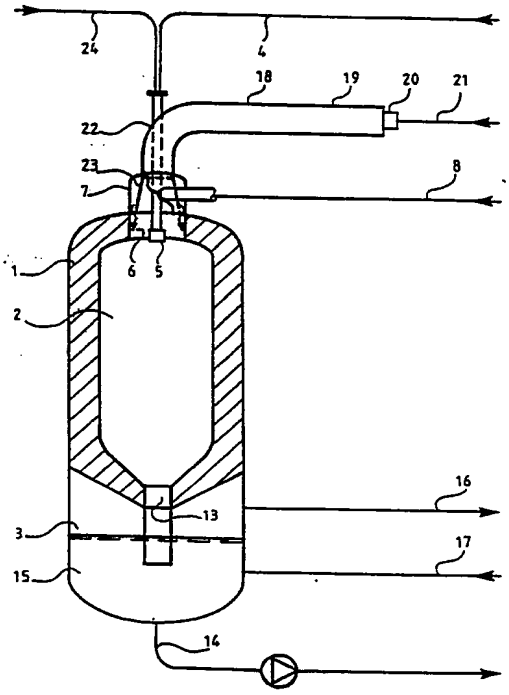
(54) Title: PROCESS IN THERMAL DECOMPOSING A CARBONACEOUS RAW MATERIAL AND REACTOR FOR CARRYING OUT THE PROCESS

(57) Abstract

A process in thermally decomposing a carbonaceous raw material in order to recover a gas having industrial value as fuel or starting material for the production of chemicals, and inorganic components in solid and/or molten form is described as well as a reactor for carrying out this process. The raw material is fed into a chamber in the reactor and the thermal decomposition is carried out at a pressure of from atmospheric pressure up to about 150 bar and at a temperature in the range of 500-1600 °C. According to the invention the thermal decomposition is carried out during exposure to low frequency sound and without or with the supply of oxygen or a gas containing oxygen in an amount below that stoichiometrically required for complete combustion of the raw material.



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Process in thermal decomposing a carbonaceous raw material and reactor for carrying out the process

The present invention relates to a process in thermally decomposing a carbonaceous raw material in order to recover a gas having industrial value as fuel or starting material for the production of chemicals, and inorganic components in solid or molten form, said raw material being fed into a chamber and the thermal decomposition being carried out at a pressure of from atmospheric pressure up to about 150 bar and at a temperature in the range of 500°-1600°C. The invention also relates to a reactor for carrying out this process.

Burning fossil fuels such as coal, oil and natural gas constitutes a large part of the production of power and heat. However, non-fossil fuels such as peat and biomass are also used, in the latter case primarily wood left over from tree felling. Production may also be based on combustion of the gas obtained upon gasification of the above-mentioned raw material. Although gasification constitutes an extra step in the production of power or heat it results in considerable advantages from the environmental and effiency points of view, which justify this extra step. Great efforts are being made to optimize the gasification process, making it even more efficient and competitive. However, the criteria for optimizing the process of gasifying a raw material, i.e. thermal decomposition with under-stoichiometric addition of oxygen, differ considerably from those applicable in combustion. This also applies if the gasification process includes a recovery of inorganic chemicals from the raw material, which is very difficult to optimize.

In some cases the termal decomposition of the raw material with under-stoichiometric addition of oxygen results in such a chemical composition of the inorganic

ash substance that it can be recovered and re-used instead of becoming a pollutant and thereby causing serious environmental problems as is often the case with combustion. The organic portion of the raw material is gasified substantially to CO, CO_2, H_2 and CH_4, whereby the residue obtained consists substantially of the inorganic components of the raw material in solid or molten form.

In such a gasification a residue of ungasified carbon is normally obtained the quantity of which being dependent upon several factors such as reaction time, reaction temperture, oxygen/raw material ratio and feeding technique. In gasification with too high under-stoichiometric conditions, the reaction temperature will normally be insufficient to achieve complete conversion of the organic substances of the raw material to combustible gas, which results in the formation of an unacceptably large amount of ungasified carbon residue. Technically, the design of the apparatus and process for the reactor is essential in particular with respect to pressure, temperature, reaction time, turbulence and the atomization of the raw material. The low oxygen/raw material ratio results in less gas generation as compared with a complete combustion process, thereby also causing difficulty in maintaining high turbulence in the reaction zone. Flow conditions with the formation of laminar layers of oxidation air and gaseous reaction products around the particles easily occur and optimization of the parameters mentioned above becomes very difficult. At higher pressure in the gasification chamber the density of the gas increases, thus causing further limitations of the possibility of achieving vigorous turbulence in the gasification chamber.

The conditions for optimizing a combustion process and those for optimizing a gasification process are thus

highly different. Combustion implies complete oxidation of the organic components of the fuel. Combustion of fuel occurs necessarily with a certain excess of air and thus differs essentially from a thermal decomposition process which occurs with limited, i.e. under-stoichiometric sypply of air (gasification) or without supply of air at all (pyrolysis). In the latter case heat must be supplied externally.

The object of the present invention is to improve the thermal decomposition technique so that the reaction processes are intensified in the greatest possible under-stoichiometric conditions, and so that gas is then generated at the highest possible conversion of the content of organic components of the raw material to gas having industrial value as fuel or as starting material for the chemical industry.

This object is achieved according to the invention in that the thermal decomposition of the raw material is carried out during exposure to low frequency sound and without or with the supply of oxygen or a gas containing oxygen in an amount below that stoichiometrically required for complete combustion of the raw material.

The raw material is suitably supplied in finely divided form. The temperature range 500°-1600°C stated in the introduction refers to the temperature of the gas at the outlet of the chamber.

Exposure to low frequency sound results in a number of valuable effects and advantages such as; (a) advanced macro and micro turbulences, which break down the laminar layers around the particles and thus constantly create new reaction surfaces for the actual diffusion-controlled reaction processes, (b) intensification of the slowest reaction step, viz. final oxidation of the residual coke,

(c) maximum carbon conversion at the lowest possible temperature, and (d) production of gas with the highest possible energy content.

The invention is described in more detail in the following, with reference to the accompanying drawing showing schematically one embodiment of a gasification reactor for carrying out the process according to the invention. The process described as an example refers to a thermal decomposition process with under-stoichiometric supply of oxygen.

With reference to the drawing the reference numeral 1 designates a vertical gasification reactor containing a brick-lined gasification chamber 2 and a cooling chamber 3 located below it. A conduit 4 for the supply of raw material is connected to the top of the reactor, the inlet 5 thereof being located inside the gasification chamber. An atomizing medium such as water steam or gas, e.g. air or oxygen gas, is supplied through a conduit 24 which is combined with the raw-material conduit 4 so that it terminates with its orifice at the same point as the conduit 4. The reactor 1 is provided with a top opening 6 which is closed by a protruding portion 7 through which said raw-material conduit 4 extends. A conduit 8 for the supply of oxidation gas, such as air, oxygen-enriched gas or pure oxygen, is connected to the casing of portion 7. This gas may suitably be pre-heated to a predetermined temperature. The gasification chamber 2 has an outlet 13 ending in the cooling chamber 3. A conduit 14 for discharging any chemical solution 15 produced is connected to the bottom of the reactor, and a conduit 16 for discharging gas recovered is connected to the reactor at a point within the cooling chamber 3 located above the liquid level therein. A conduit 17 is also connected to the bottom portion of the reactor for the addition of liquid and circulation of any chemical solution formed.

The method described above for cooling the gas from the reactor and separating inorganic ash material is only one of several possible methods.

The gas obtained from the gasification is conveyed to a cleaning step and can thereafter be utilized for producing energy or as starting material in the chemical industry, e.g. for the production of ammonia, methanol and synthetic natural gas.

According to the present invention the rector also includes a sound generator means 18 which generates and maintains a low frequency sound in the gasification chamber 2 so that the atomized raw material injected through inlet 5, the gas present, and the decomposed substances are subjected to the influence of low frequency sound. In the embodiment shown, said sound generator means comprises a tubular resonator 19, the length of which is suitably one fourth of the wave length of the sound generated, and a supply unit 20 disposed at one end of the resonator and forming a low frequency generator. The supply unit 20 is connected to a conduit 21 for the supply of propellant gas such as air. The generator may consist of an infra-sound generator, e.g. of the type described in US 4,593,962. However, any type of low frequency sound generator means can be used for the purpose of the invention. The resonator 19 describes a 90° bend 22 and terminates in a diffusor 23 which, together with the bend 22, is included in the quarter wave generator. The diffusor 23 is located inside and is surrounded by the casing of said portion 7. The low frequency generator may be located at other places than that shown, e.g. on the side walls or at the bottom of the gasification chamber 2. If desired the reactor may be provided with several sound generators supplying low frequency sound at a different points in the gasification chamber 2.

The high reflection ability of infra-sound enables a large space to be filled with sound from a single infra-sound generator. No sound shadows occur and the sound level remains unchanged and occurs simultaneously in all parts of the gasification chamber. The reactor includes control equipment (not shown) which maintains the entire system - resonance tube and gasification chamber - in resonance even when the operating conditions change such as at a change in temperature and pressure. The infra-sound oscillates the gas and the particles suspended therein by cyclic compressions and decompressions, thus breaking down the laminar gas layers around the particles and thereby enabling a considerable increase in the contact between the suspended particles and the surrounding gas, due to the well developed macro as well as micro turbulence, resulting from the influence of the low frequency sound. New attack points are thus constantly created for chemical reactions.

The use of low frequency sound gives essential improvements in the gasification system described, involving improved transport of reaction substances by means of low frequency sound. The velocity of the oxygen molecule on its way to the particles of raw material containing carbon and other organic substances is dependent on the diffusion resistance in the laminar gas layer closest to the surface of each particle. The reaction rate is thus increased by the vigorous turbulence in the laminar layer produced by the process according to the invention. The slowest step in the gasification process is the final oxidation of the residual coke. This step is also controlled by the transport of oxygen and water steam molecules through the laminar gas layer surrounding the particles. Reactants in the gas phase must thus pass into the glowing coke particle through the layer of gas surrounding it. The degree of carbon conversion is thus improved by means of

the method described above. The description above thus applies specifically in the case of under-stoichiometric supply of oxygen, i.e. gasification.

The thermal decompostion takes place in the apparatus described above during controlled supply of oxygen or a gas containing oxygen in an amount below that stoichiometrically required for complete oxidation of the raw material. This amount of oxygen corresponds to about 20-80%, preferably about 30-60%, of the amount of oxygen stoichiometrically required for complete oxidation. Alternatively the thermal decomposition is performed as a pyrolysis, i.e. without the addition of oxygen. The pressure in the reactor may be from atmospheric pressure up to 150 bar (abs), preferably 15-35 bar (abs).

The inorganic components obtained may consist of or contain chemicals of industrial value depending on the type of raw material which is thermally decomposed. An example of such raw material is spent liquor from the production of pulp. When gasifying sulphate spent liquors obtained in the cellulose industry, for instance, both the organic and the inorganic substances are to be recovered, viz. both the wood lignin released during sulphate digestion, which is recovered in the form of an energy-rich gas, and also digestion chemicals in the form of sodium and sulphur compounds. The sulphur shall be recovered in sulphide form and the process requires high under-stoichiomtric (reducing) conditions within a temperature range that depends on which method is used for gasifying the spent liquor. There are presently the dry method which operates within the temperature range of 500°-800°C and involves substantially that the sodium forms Na_2CO_3 in solid form and the sulphur forms H_2S in the gas produced; the melt method which operates within the temperature range of 800°-1000°C and involves that molten droplets of Na_2CO_3 and Na_2S are formed as well as

a gas containing minor amounts of H_2S; and the causticizing-free melt method which operates within the temperature range of 1000°-1500°C and results in direct conversion of inorganic material to active digestion chemicals in the form of Na_2S and NaOH. The chemicals recovered are re-used in the pulp production process.

In a comparative test the thermal decomposition was carried out first without and then with exposure to low frequency sound having a frequency of 20 Hz in a reactor of the design described above. In other respects the reaction conditions were identical, viz. a temperature of 910°C and a pressure of 0.5 bar overpressure, the raw material being concentrated black liquor atomized in identical manner. Exposure to low frequency sound of the stated frequency resulted in an 11% improvement of the degree of carbon conversion and an associated 8% increase in the amount of energy in the gas produced. Additional tests showed that the same degree of carbon conversion and amount of energy in the gas produced could be achieved at a reaction temperature which was 75°C lower when gasification was carried out under exposure to low frequency sound with respect to gasification without such sound.

C L A I M S

1. A process in thermally decomposing a carbonaceous raw material in order to recover a gas having industrial value as fuel or starting material for the production of chemicals, and inorganic components in solid and/or molten form, said raw material being fed into a chamber and the thermal decomposition being carried out at a pressure of from atmospheric pressure up to about 150 bar and at a temperature in the range of 500°-1600°C, characterized in that the thermal decomposition is carried out during exposure to low frequency sound and without or with the supply of oxygen or a gas containing oxygen in an amount below that stoichiometrically required for complete combustion of the raw material.

2. A process according to claim 1, characterized in that the low frequency sound has a frequency of at most 150 Hz, preferably at most 40 Hz and most preferably at most 20 Hz.

3. A process according to claim 1 or 2, characterized in that oxygen or gas contantaining oxygen is supplied in an amount corresponding to about 20-80%, preferably 30-60%, of the stoichiometrical amount for complete combustion of the raw material.

4. A process according to any of claims 1-3, characterized in that the low frequency sound is supplied in close connection to the location for the supply of the raw material.

5. A process according to any of claims 1-4, characterized in that the low frequency sound is generated by at least one sound generator means (18).

6. A process according to any of claims 1-5, characterized in that the raw material and the oxidation gas if any, is/are preheated to a predetermined temperature.

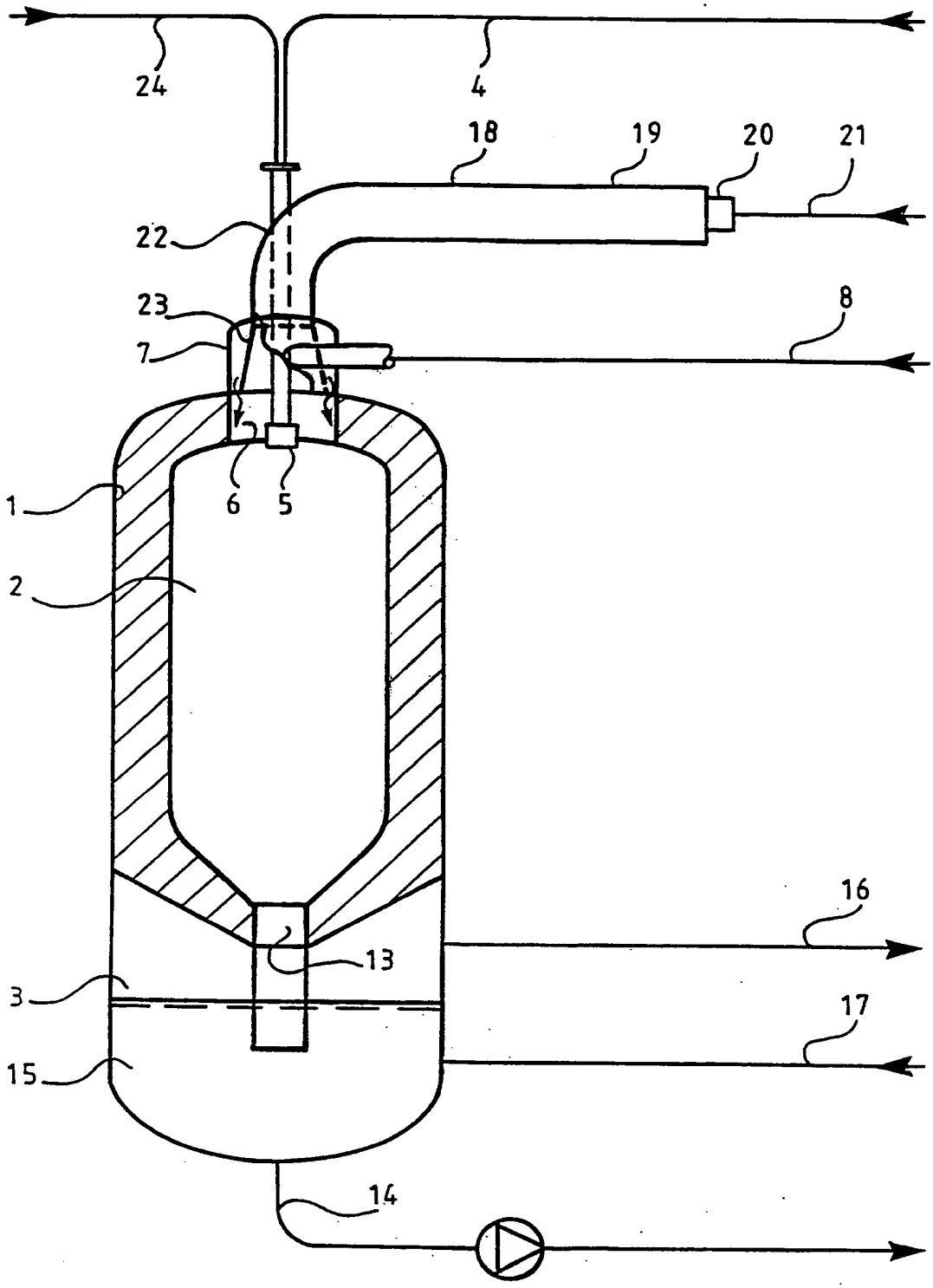
7. A process according to any of claims 1-6, characterized in that the raw material used is spent liquor consisting of residual products containing sodium and sulphur from the production of sulphate pulp or of residual products from the production of sulphite pulp.

8. A reactor for carrying out the process according to any of claims 1-7 for thermally decomposing a carbonaceous raw material in order to recover a gas having industrial value as fuel or starting material for the production of chemicals, and inorgaic components in solid and/or molten form, said raw material being supplied into a chamber and the thermal decomposition being carried out at a pressure of from atmospheric pressure up to about 150 bar and at a temperature in the range of about 500°-1600°C, characterized in at least one sound generator means (18) arranged to generate low frequency sound in the reactor.

9. A reactor according to claim 8, characterized in that the sound generator means (18) is arranged to generate low frequency sound of a frequency of at most 150 Hz, preferably at most 40 Hz and most preferably at most 20 Hz.

10. A reactor according to claim 8 or 9, characterized in that the sound generator means (18) is arranged to supply low frequency sound to the reactor in close connection to the location for the supply of the raw material.

11. A reactor according to any of claims 8-10, characterized in that the low frequency sound is generated by at least one sound generator means (18).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INTERNATIONAL SEARCH REPORT

International Application No PCT/SE 91/00075

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all)[6]

According to International Patent Classification (IPC) or to both National Classification and IPC

IPC5: C 10 J 3/00, C 10 B 53/02, D 21 C 11/12, F 23 G 5/027

II. FIELDS SEARCHED

Minimum Documentation Searched[7]

Classification System	Classification Symbols
IPC5	C 10 B; C 10 J; F 23 G

Documentation Searched other than Minimum Documentation to the Extent that such Documents are Included in Fields Searched[8]

SE,DK,FI,NO classes as above

III. DOCUMENTS CONSIDERED TO BE RELEVANT[9]

Category *	Citation of Document,[11] with indication, where appropriate, of the relevant passages[12]	Relevant to Claim No.[13]
A	Derwent's Abstract, No. 82-23747E/12, SU 834 373, A, 810531, publ. week 8212, Dialog Information Service, File 351, WPI --	1
A	SE, B, 458799 (INSAKO AB) 8 May 1989, see abstract; figure 1	1-7
X	--	8-11
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IV. CERTIFICATION

Date of the Actual Completion of the International Search	Date of Mailing of this International Search Report
30th April 1991	1991 -05- 02
International Searching Authority	**Signature of Authorized Officer**
SWEDISH PATENT OFFICE	Jan Carlerud

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ANNEX TO THE INTERNATIONAL SEARCH REPORT ON INTERNATIONAL PATENT APPLICATION NO. PCT/SE 91/00075

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Patent document cited in search report	Publication date	Patent family member(s)		Publication date
SE-B- 458799	89-05-08	AU-B-	569561	88-02-04
		AU-D-	3607684	85-06-06
		CA-A-	1237650	88-06-07
		EP-A-B-	0144918	85-06-19
		JP-A-	60211214	85-10-23
		SE-A-	8306653	85-06-03
		SU-A-	1452494	89-01-15
		US-A-	4650413	87-03-17